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(Patent)

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Patent Application of:
Yoshiki HASHIZUME et al.

Application No.:	10/525,068	Confirmation No.:	5831
Filed:	February 18, 2005	Art Unit:	1793
For:	ALUMINUM PIGMENT, METHOD OF MANUFACTURING THE SAME AND RESIN COMPOSITION	Examiner:	S. Abu Ali

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Yoshiki Hashizume, declare and say as follows:

I graduated from Kyoto University, Faculty of Engineering, in March 1975. Since April 1976, I have been employed by Toyo Aluminium Kabushiki Kaisha, engaged in research and development of aluminum materials (aluminum paste, aluminum powder, aluminum nitride powder, etc.). Currently, I am a Manger, Research & Development Dept., Core Technology Center.

I am familiar with U.S. Application Serial No. 10/525,068, of which I am a co-inventor. The following experiments were conducted either by myself or under my direct supervision, and show the superior results obtained by the present invention.

PREPARATION OF SAMPLES

Experiment 1 (corresponding to Example 10 in the present application)

A solution was obtained by gradually adding 0.5 g of metallic molybdenum powder to 10 g of hydrogen peroxide water containing 30% of hydrogen peroxide and reacting the same. The solution was dissolved in 600 g of isopropyl alcohol, and stirred at 50°C for 1 hour with addition of 153.8 g (aluminum content: 100 g) of a commercially available aluminum pigment (7640NS by Toyo Aluminium K.K., solid content: 65 %).

Thereafter monoethanolamine was added in order to adjust the pH value of the slurry to 8.5.

Then, 30 g of tetraethoxysilane (hereinafter abbreviated as TEOS) and 10 g of phenyl triethoxysilane were added to the pH-adjusted slurry, which in turn was further stirred at 50°C for 10 hours. The pH value of the slurry was checked every 2 hours in this process, and adjusted to be 8.5 by adding monoethanolamine.

After termination of the aforementioned reaction, the slurry was solid-liquid separated through a filter, and the slurry containing the obtained aluminum pigment was dried at 105°C for 3 hours to obtain a powdered aluminum pigment.

Experiment 2 (Comparative Example)

A sample was prepared in a similar way to Experiment 1, except that no metallic molybdenum powder was added.

Experiment 3 ***(Comparative Example)***

A sample was prepared in a similar manner to Experiment 1, except that 0.01g of metallic molybdenum powder was used.

Experiment 4 ***(Example)***

A sample was prepared in a similar manner to Experiment 1, except that 0.02 g of metallic molybdenum powder was used.

Experiment 5 ***(Example)***

A sample was prepared in a similar manner to Experiment 1, except that 50g of hydrogen peroxide water, and 4.0g of metallic molybdenum powder were used.

Experiment 6 ***(Comparative Example)***

A sample was prepared in a similar manner to Experiment 1, except that 70g of hydrogen peroxide water and 6.0g of metallic molybdenum powder were used.

Experiment 7 ***(Comparative Example)***

A sample was prepared in a similar manner to Experiment 1 except 4.5g of TEOS was used.

Experiment 8 ***(Example)***

A sample was prepared in a similar manner to Experiment 1, except that 8.0g of TEOS was used.

Experiment 9 ***(Example)***

A sample was prepared in a similar manner to Experiment 1, except that 180g of TEOS was used.

Experiment 10 ***(Comparative Example)***

A sample was prepared in a similar manner to Experiment 1, except that 200g of TEOS was used.

EVALUATION OF SAMPLES

1. *Measurement of Si and Mo Content*

The content of Mo and Si in each aluminum pigment were determined by plasma spectral analysis with a ICPS-8000 device (manufactured by Shimadzu Corporation) through a calibration curve employing a liquid extracted by an alkali dissolution/extraction method and Mo and Si standard solutions.

2. *Samples' Stability with Time*

100 parts by mass of water was added to 100 parts by mass of powder of each aluminum pigment. The mixture was pasted, the paste was preserved at 50°C for 1 month and thereafter manually wet-screened in water with a JIS standard sieve having an aperture of 45 μm in order to evaluate the stability, as follows:

O : aluminum pigment hardly changed

Δ : aluminum pigment partially agglomerated

x : aluminum pigment mostly agglomerated

3. *Measurement of Gas Yield in Water-Based Paint*

Each aluminum pigment was used for preparing a water-based paint having the following composition:

Water-soluble acrylic resin (*1)	28.2 g
Melamine resin (*2)	4.4 g
Triethanolamine	1.1 g
Deionized water	44.8 g

Isopropyl alcohol	3.0 g
Transparent iron oxide (*3)	5.0 g
Aluminum pigment	3.0 g (solid content)

*1: Armatex WA911 by Mitsui Toatsu Chemicals, Inc.
*2: Cymel 350 by Mitsui Toatsu Chemicals, Inc.
*3: SICOTRANS RED L2175D by BASF

80 g of the water-based paint prepared in the aforementioned manner was sampled and stored in a water boiler adjusted to 50°C for 7 days, in order to measure the cumulative hydrogen gas yield by water displacement with a measuring cylinder.

4. ***Color Tone Evaluation of Film of Water-Based Paint Containing Aluminum Pigment***

Each water-based paint (prepared as described above) was air-sprayed to a test steel plate previously electrodeposition-coated with a temporary rust-prevention paint so that the thickness was 13 μm after drying and predried at 90°C for 10 minutes. An organic solvent type top coat paint having the following composition was air-sprayed so that the thickness was 40 μm after drying, and dried at 140°C for 30 minutes in order to prepare a metallic painted plate.

Acrylic resin (*4)	140 g
Melamine resin (*5)	50 g
Solvesso 100	60 g

*4: Armatex 110 by Mitsui Toatsu Chemicals, Inc.
*5: Uban 205E60 by Mitsui Toatsu Chemicals, Inc.

The metallic luster of the metallic painted plate obtained in the aforementioned manner was subjected to a 5-stage evaluation on the basis of the following standards. The term "flip

flop" refers to the variation in lightness with the angle of observation. The lightness changes as the flip flop gets stronger.

5: extremely excellent in brightness and extremely strong in flip flop

4: excellent in brightness and strong in flip flop

3: ordinary both in brightness and in flip flop

2: slightly inferior in brightness and slightly weak in flip flop

1: inferior in brightness and weak in flip flop

8. ***Film Moisture Resistance/Adhesion Test***

Each painted plate obtained as described above was left in an atmosphere of 40°C with moisture of 100% for 10 days, and an adhesion test was thereafter performed according to JIS K5600 5-6:1999. The following Tables show the obtained results. The results were evaluated as follows:

5: no peeling

4: peeling: not more than 10%

3: peeling: 10 to 50%

2: peeling: 50 to 90%

1: peeling: 100%

	Experiment 1	Experiment 2	Experiment 3	Experiment 4	Experiment 5	Experiment 6
Mo/H_2O_2 30% aq	0.5/10	0/10	0.01/10	0.02/10	4.00/50	6.00/70
Analysis Value of Sample Mo (wt% vs Al)	0.45	0	0.008	0.014	3.80	5.78
Analysis Value of Sample Si (wt% vs Al)	4.6	1.1	2.8	3.8	4.5	4.6
Stability (50°C, 1 month)	○	○	○	○	○	×
Gas Yield	0	13	3	0	0	4
Color Tone of Painted Plate	5	2	3	4	3	1
Moisture Resistance /Adhesiveness	5	3	5	5	5	2

	Experiment 7	Experiment 8	Experiment 9	Experiment 10
Mo/H_2O_2 30% aq	0.5/10	0/10	0.01/10	0.02/10
Analysis Value of Sample Mo (wt% vs Al)	0.46	0.45	0.45	0.45
Analysis Value of Sample Si (wt% vs Al)	0.69	1.25	19.8	22.3
Stability (50°C, 1 month)	○	○	○	○
Gas Yield	8	0	0	0
Color Tone of Painted Plate	5	4	3	1
Moisture Resistance /Adhesiveness	2	5	5	5

Examples: Experiments 1, 4, 5, 8, 9

Comparative Examples: Experiments 2, 3, 6, 7, 10

RESULTS

Experiments 1-6

The results of Experiments 1 to 6 show that when the Mo content is lower than that defined in claim 1 of the present application, the anchor effect is not exhibited and it is difficult to form a silica coat on the aluminum pigment. In such cases, the Si content is particularly low and gas generation cannot be suppressed, and therefore, such a sample cannot be used as an aluminum pigment for a water-based paint (see Experiments 2-3). When the Mo content is higher than that defined in claim 1 of the present application, the aluminum pigment agglomerates, resulting in lack of stability and considerable degradation of the color tone (see Experiment 6).

Experiments 7-10

The results of Experiments 7 to 10 show that when the content of Si derived from a silica coat is too low, the color tone of the painted plate is actually superior (see Experiment 7). However, this is merely because a smaller thickness of the silica coat allows a good color tone of the bare aluminum pigment itself to be exhibited, and generation of hydrogen gas in the water-based paint cannot be suppressed. When the Si content is too high, generation of hydrogen gas in the water-based paint can actually be suppressed. The silica layer, however, is too thick, which results in degradation of the color tone of the pigment itself (see Experiment 10).

Clearly, superior results are obtained at the claimed Mo and Si content.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Yoshiki Hashizume
Signature

Yoshiki HASHIZUME
Typed or Printed Name

December 3, 2010
Date